

# PATENT COOPERATION TREATY

From the  
INTERNATIONAL PRELIMINARY EXAMINING AUTHORITY

## PCT

To:

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10 FEB. 2005

### NOTIFICATION OF TRANSMITTAL OF THE INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Rule 71.1)

Date of mailing  
(day/month/year)

08.02.2005

Applicant's or agent's file reference  
Cal 86764

#### IMPORTANT NOTIFICATION

International application No.  
PCT/EP 03/10937

International filing date (day/month/year)  
26.09.2003

Priority date (day/month/year)  
27.09.2002

Applicant  
ENI S.P.A.

1. The applicant is hereby notified that this International Preliminary Examining Authority transmits herewith the international preliminary examination report and its annexes, if any, established on the international application.
2. A copy of the report and its annexes, if any, is being transmitted to the International Bureau for communication to all the elected Offices.
3. Where required by any of the elected Offices, the International Bureau will prepare an English translation of the report (but not of any annexes) and will transmit such translation to those Offices.
4. **REMINDER**

The applicant must enter the national phase before each elected Office by performing certain acts (filing translations and paying national fees) within 30 months from the priority date (or later in some Offices) (Article 39(1)) (see also the reminder sent by the International Bureau with Form PCT/IB/301).

Where a translation of the international application must be furnished to an elected Office, that translation must contain a translation of any annexes to the international preliminary examination report. It is the applicant's responsibility to prepare and furnish such translation directly to each elected Office concerned.

For further details on the applicable time limits and requirements of the elected Offices, see Volume II of the PCT Applicant's Guide.

The applicant's attention is drawn to Article 33(5), which provides that the criteria of novelty, inventive step and industrial applicability described in Article 33(2) to (4) merely serve the purposes of international preliminary examination and that "any Contracting State may apply additional or different criteria for the purposes of deciding whether, in that State, the claimed inventions is patentable or not" (see also Article 27(5)). Such additional criteria may relate, for example, to exemptions from patentability, requirements for enabling disclosure, clarity and support for the claims.

Name and mailing address of the international preliminary examining authority:



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# PATENT COOPERATION TREATY

## PCT

### INTERNATIONAL PRELIMINARY EXAMINATION REPORT (PCT Article 36 and Rule 70)

Applicant's or agent's file reference Cal 86764	<b>FOR FURTHER ACTION</b> See Notification of Transmittal of International Preliminary Examination Report (Form PCT/PEA/416)	
International application No. PCT/EP 03/10937	International filing date ( <i>day/month/year</i> ) 26.09.2003	Priority date ( <i>day/month/year</i> ) 27.09.2002
International Patent Classification (IPC) or both national classification and IPC C10G27/12		
Applicant ENI S.P.A.		

<p>1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.</p> <p>2. This REPORT consists of a total of 10 sheets, including this cover sheet.</p> <p><input checked="" type="checkbox"/> This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).</p> <p>These annexes consist of a total of 2 sheets.</p>
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<p>3. This report contains indications relating to the following items:</p> <ul style="list-style-type: none"> <li>I <input checked="" type="checkbox"/> Basis of the opinion</li> <li>II <input type="checkbox"/> Priority</li> <li>III <input type="checkbox"/> Non-establishment of opinion with regard to novelty, inventive step and industrial applicability</li> <li>IV <input type="checkbox"/> Lack of unity of invention</li> <li>V <input checked="" type="checkbox"/> Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement</li> <li>VI <input type="checkbox"/> Certain documents cited</li> <li>VII <input type="checkbox"/> Certain defects in the international application</li> <li>VIII <input type="checkbox"/> Certain observations on the international application</li> </ul>
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Date of submission of the demand  03.03.2004	Date of completion of this report  08.02.2005
Name and mailing address of the international preliminary examining authority:  <div style="display: flex; align-items: center;"> <div>             European Patent Office - P.B. 5818 Patentlaan 2              NL-2280 HV Rijswijk - Pays Bas              Tel. +31 70 340 - 2040 Tx: 31 651 epo nl              Fax: +31 70 340 - 3016           </div> </div>	Authorized Officer  Gilliquet, J-N  Telephone No. +31 70 340-4573



**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT**

International application No. **PCT/EP 03/10937**

**I. Basis of the report**

1. With regard to the **elements** of the international application (*Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17)*):

**Description, Pages**

1-35 as originally filed

**Claims, Numbers**

1-43, 45, 47-49 as originally filed  
44, 46 received on 08.11.2004 with letter of 05.08.2004

**Drawings, Sheets**

1/1 as originally filed

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language: , which is:

- ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).  
☐ the language of publication of the international application (under Rule 48.3(b)).  
☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- ☐ contained in the international application in written form.  
☐ filed together with the international application in computer readable form.  
☐ furnished subsequently to this Authority in written form.  
☐ furnished subsequently to this Authority in computer readable form.  
☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.  
☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

- ☐ the description, pages:  
☐ the claims, Nos.:  
☐ the drawings, sheets:

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT**

International application No. **PCT/EP 03/10937**

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5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)).

*(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)*

6. Additional observations, if necessary:

**V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**

1. Statement

Novelty (N)	Yes: Claims	1-33,36,37,43-49
	No: Claims	34,35,38-42
Inventive step (IS)	Yes: Claims	1-30,43-49
	No: Claims	31-42
Industrial applicability (IA)	Yes: Claims	1-49
	No: Claims	

2. Citations and explanations

**see separate sheet**

**Re Item V**

**Reasoned statement with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**

Reference is made to the following documents:

- D1: WO 02 053683 A (CARRIAT JEAN-YVES ;TOTAL RAFFINAGE DISTRIB S A (FR); BISSON MARC ( ) 11 July 2002 (2002-07-11)
- D2: WO 01 48119 A (BERNARD JEAN RENE ;FAJULA FRANCOIS (FR); RABION ALAIN (FR); ELF AN) 5 July 2001 (2001-07-05)
- D3: US-A-5 049 536 (BELLUSSI GIUSEPPE ET AL) 17 September 1991 (1991-09-17)
- D4: US-B1-6 323 147 (TSUJI JUNPEI ET AL) 27 November 2001 (2001-11-27)

**1. Amendments**

The amendments filed with letter dated 5.11.2004 to the International Bureau under Article 34(2)(a) do not introduce subject-matter which extends beyond the content of the application as filed, in accordance with Article 34(2)(b) PCT.

**2. Lack of support**

Independent claims 1, 31-34, 43, 45 and 48 are not supported by the description as required by Article 6 PCT, as their scope is broader than justified by the description and examples.

Claim 1 refers to mixed oxide containing an oxide matrix selected from silica, alumina, ceria, magnesia and mixtures thereof, wherein one or more oxidative metal oxides selected from transition metal oxides and group IVA metal oxides are uniformly dispersed.

Claims 31-34, 43, 45 and 48 refer also to other mixed oxides with diverse lists of elements.

On the other hand, the description discloses examples only with mixtures of silica and titania.

3. Clarity

The terms "micro and/or mesoporous" and "micro-mesoporous" used in independent claims 1, 31-34, 43, 45 and 48 are vague and unclear: are macropores absent in the oxide ? Is there a bimodal porosity ?

These terms leave the reader in doubt as to the meaning of the technical features to which they refer, thereby rendering the definition of the subject-matter of said claims unclear, Article 6 PCT.

From the description it appears that the average pore diameter is between 2.4 and 3.7 nm.

4. Novelty and inventive step of present independent claim 1 and dependent claims 2-30

The objections of points 2 and 3 above notwithstanding, it would appear that the subject-matter of present independent claim 1 could be considered as new and inventive.

Document D1 (See claims 1-12 and examples 1-2 of D1) is regarded as being the closest prior art to the subject-matter of claim 1. It discloses a process for oxidative desulphurization of hydrotreated hydrocarbon mixtures which boil within the range of 180 °C to 360 °C, containing less than 350 ppm of sulphur as thiophenic compounds in the presence of an organic peroxide and a catalyst having a silica matrix, impregnated with WO<sub>3</sub> or MoO<sub>3</sub> (See example 1).

The subject-matter of claim 1 differs from this known process in that the catalyst is made of a mixture of oxides uniformly dispersed, not of an oxide impregnated on another oxide.

The subject-matter of claim 1 is therefore new (Article 33(2) PCT).

The technical effect of this distinguishing feature is an alternative process.

The problem to be solved by the present invention may therefore be regarded as how to provide an alternative process.

None of the documents of the prior art on file discloses such an alternative process nor gives a hint to its possibility, so that it can be considered as inventive in the sense of Art. 33 (3) PCT.

Claims 2-30 are dependent on claim 1 and as such also meet the requirements of the PCT with respect to novelty and inventive step.

5. Novelty and inventive step of present independent claim 31

The objections of points 2 and 3 above notwithstanding, it would appear that the subject-matter of present independent claim 31 could be considered as new but not as inventive.

Document D3 (See claim 1 and col.3 l.20-47 of D3) is regarded as being the closest prior art to the subject-matter of claim 31. It discloses catalytic compositions comprising a completely amorphous micro-mesoporous mixed oxide having a molecular ratio of  $\text{SiO}_2 / \text{Al}_2\text{O}_3$  comprised within the range of from 30/1 to 500/1, a surface area comprised within the range of from 500 to 1000  $\text{m}^2/\text{g}$ , a total volume of pores comprised within the range of from 0.3 to 0.6  $\text{ml/g}$ , an average diameter of the pores of the order of magnitude of 10  $\text{\AA}$  or less and including oxides of titanium or zirconium as binder.

The subject-matter of claim 31 differs from this known composition in that the catalyst is made of a mixture of oxides uniformly dispersed and not of an oxide mechanically mixed as a binder.

The subject-matter of claim 31 is therefore new (Article 33(2) PCT).

In the absence of examples showing a technical effect of this distinguishing feature, this subject-matter can not be considered as inventive because it provides an obvious alternative to a known composition.

Moreover, the examples of the application do relate to silica and not to silica-alumina, so that no technical effect of the claimed invention is provided.

Therefore the subject-matter of present independent claim 31 can not be considered as inventive in the sense of Art. 33 (3) PCT.

6. Novelty and inventive step of present independent claim 32

The objections of points 2 and 3 above notwithstanding, it would appear that the subject-matter of present independent claim 32 could be considered as new but not as inventive.

Document D1 (See claim 1 and example 1 of D1) is regarded as being the closest prior art to the subject-matter of claim 32. It discloses catalytic compositions comprising a completely amorphous micro-mesoporous mixed oxide consisting of a matrix of silica impregnated with WO<sub>3</sub> or MoO<sub>3</sub> (See example 1).

The subject-matter of claim 32 differs from these known compositions in that the catalyst is made of a mixture of oxides, not of an oxide impregnated on another oxide. Moreover the metal oxides uniformly dispersed are different.

The subject-matter of claim 32 is therefore new (Article 33(2) PCT).

But the examples of the application relate only to catalyst compositions comprising silica-titania, which are subject of the disclaimer of present independent claim 32, meaning that only examples falling into the scope of the disclaimer are provided in present application. Therefore no technical effect of the distinguishing feature is established.

Therefore the subject-matter of present independent claim 32 can not be considered as inventive in the sense of Art. 33 (3) PCT.

7. Novelty and inventive step of present independent claim 33

The objections of points 2 and 3 above notwithstanding, it would appear that the subject-matter of present independent claim 33 could be considered as new but not as inventive.



Document D1 (See claims 1 and 6 and example 1 of D1) is regarded as being the closest prior art to the subject-matter of claim 33. It discloses catalytic compositions comprising a completely amorphous micro-mesoporous mixed oxide consisting of a matrix of silica impregnated with WO<sub>3</sub> or MoO<sub>3</sub> (See example 1).

The subject-matter of claim 33 differs from these known compositions in that the catalyst is made of a mixture of oxides, not of an oxide impregnated on another oxide. Moreover the metal oxides uniformly dispersed are different.

The subject-matter of claim 33 is therefore new (Article 33(2) PCT).

But the examples of the application relate only to catalyst compositions comprising silica-titania of the type MSA or ERS-8, which are subject of the disclaimer of present independent claim 33, meaning that only examples falling into the scope of the disclaimer are provided in present application. Therefore no technical effect of the distinguishing feature is established.

Therefore the subject-matter of present independent claim 33 can not be considered as inventive in the sense of Art. 33 (3) PCT.

8. Novelty and inventive step of present independent claim 34 and dependent claims 35-42

The objections of points 2 and 3 above notwithstanding, it would appear that the subject-matter of present independent claim 34 could not be considered as new.

Document D4 (See comparative example 1 of D4) discloses catalytic compositions comprising a completely amorphous micro- and/or mesoporous mixed oxides containing a matrix of silica, wherein oxides of titanium are uniformly dispersed, whose surface has -O-Si(R)<sub>3</sub> groups. The subject-matter of present independent claim 34 can therefore not be considered as new in the sense of Article 33(2) PCT.

The subject-matter of present dependent claims 35 and 38-42 of the present application is disclosed in document D4, so that it can not be considered as new. Therefore it does not satisfy the criterion set forth in Article 33(2) PCT.

From the text of the description of the present application, the subject-matter of

dependent claims 36 and 37 does not contribute to solving the technical problem posed in the application. Therefore it does not involve an inventive step in the sense of Article 33(3) PCT.

9. Novelty and inventive step of present independent claims 43, 45 and 48 and dependent claims 44, 46, 47 and 49

The objections of points 2 and 3 above notwithstanding, it would appear that the subject-matter of present independent claims 43, 45 and 48 could be considered as new and inventive.

Document D4 (See claim 1, col.4 §9, col.4 last § -col.5 last § and comparative example 1 of D4), which is considered to be the closest prior art, discloses a process for preparing the catalytic compositions similar to the one claimed in present independent claims 43, 45 and 48.

The subject-matter of these claims differs from this known process in that the ammonium cations used are different and the gel obtained is calcined.

The subject-matter of these claims is therefore new (Article 33(2) PCT).

The technical effect of these distinguishing features is an alternative process.

The problem to be solved by the present invention may therefore be regarded as how to provide an alternative process.

None of the documents of the prior art on file discloses such an alternative process nor gives a hint to its possibility, so that it can be considered as inventive in the sense of Art. 33 (3) PCT.

Claims 44, 46, 47 and 49 are dependent on these claims and as such also meet the requirements of the PCT with respect to novelty and inventive step.

10. Further clarity and conciseness objections

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT - SEPARATE SHEET**

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International application No. PCT/EP 03/10937

Although claims 31, 32, 33 and 34 have been drafted as separate independent claims, they appear to relate effectively to the same subject-matter and to differ from each other only with regard to the definition of the subject-matter for which protection is sought. The aforementioned claims therefore lack conciseness and as such do not meet the requirements of Article 6 PCT.

Although claims 43, 45 and 48 have been drafted as separate independent claims, they appear to relate effectively to the same subject-matter and to differ from each other only with regard to the definition of the subject-matter for which protection is sought. The aforementioned claims therefore lack conciseness and as such do not meet the requirements of Article 6 PCT.

Butyl > 73

Propyl > 53

operating at a temperature close to the boiling point,  
at atmospheric pressure, of the alcohol used and of any  
5 alcohol which develops as by-product of the above  
hydrolysis reaction, without the elimination or  
substantial elimination of said alcohols from the  
reaction environment, preferably at a temperature of  
between 20°C and 80°C;

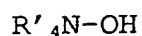
10 (b) subjecting the gel obtained in step (a) to  
drying;

(c) outgassing the obtained material in vacuum and then  
adding a solution of silylating agent in solvent;

(d) refluxing the resulting mixture under inert  
15 atmosphere and then filtering the obtained product,  
washing and subjecting it to drying.

44) Process <sup>according to</sup> ~~for preparing the catalytic composition~~  
~~of~~ claim 43 comprising :

(a) subjecting to hydrolysis and gelification a  
20 solution of one or more soluble or hydrolyzable  
compounds of Si and Al, in alcohol, with an aqueous  
solution of a hydroxide of tetra-alkylammonium having  
the formula (I):



accordance with the values shown in table B below :

Table B

	R'	H <sub>2</sub> O/R' <sub>4</sub> N <sup>+</sup>
	Hexyl	≤ 133
5	Pentyl	≤ 100
	Butyl	≤ 73
	Propyl	≤ 53

operating at a temperature close to the boiling point,  
at atmospheric pressure, of the alcohol used and of any  
10 alcohol which develops as by-product of the above  
hydrolysis reaction, without the elimination or  
substantial elimination of said alcohols from the  
reaction environment, preferably at a temperature of  
between 20°C and 80°C;

15 (b) subjecting the gel obtained in step (a) to  
drying;

(c) the obtained material is outgassed in vacuum and  
then a solution of silylating agent in solvent;

(d) refluxing the resulting mixture under inert  
20 atmosphere and then filtering the obtained product,  
washing and subjecting it to drying.

<sup>according to</sup>  
46) Process ~~for preparing the catalytic composition~~  
of claim 45 comprising :